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A semiclassical model for current-induced stress in electromigration and electroplasticity

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Abstract

We derive and employ a new semiclassical model to interpret the mechanism of the current-induced stress. We assume the metal clusters in spherical jellium model as hard sphere and use spherical flow model in classical fluid mechanics and quantum viscosity to calculate the cluster radius. The results indicate that there might be large-group transportation but not only single-particle transport in electromigration. Two new kinds of current-induced stress are derived based on present model, which are quantum density stress and quantum viscous stress. These two kinds of quantum stress provide new ways to estimate the current-induced stress. Our model is also suitable for the motion of nanoisland and void. Our model provides new sight into applying hydrodynamics to electromigration.

Introduction.—Passage of large current in a metal can generate preferential diffusion of atoms in one direction. This preferential diffusion of atoms builds up compressive stresses near the anode due to the addition of atoms, whereas tensile

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stresses are found near the cathode due to the removal of material.[1-3] The mechanical stress leads to grain boundary and interface diffusion, which is known to be driven by electron wind force.[4-6] The effect of current-induced stress in atomic and molecular wires has been studied in recent studies.[7-17] Previous experiments have found that electromigration can have a profound influence upon the development of surface morphology and material plasticity.[4-10] The mechanism of electron wind force had also been broadly studied.[11-15] However, some important properties of conducting electrons failed to be considered, such as the quantum viscosity. Also, some phenomenon cannot be well explained, such as the grain boundary drag effect,[16] the motion of nanoislands.[17]

In this article, we provide a new way to interpret the mechanism of current-induced stress. We assume the conduction electrons as electron fluid. We mainly use the spherical jellium model,[18,19] the quantum Bernoulli equation[20], the quantum viscosity[21], and the classical spherical flow model in fluid mechanics[22]. According to the spherical jellium model, nearspherical metal clusters with specific number of valence electrons, e.g., 8, 18, 20, 34, 40, 58 are very strong because of their enclosed electronic shells.[18,19] These clusters are called magic-number clusters due to their exceptional stability.[23,] The jellium model had been used to analyze the grain boundary drag effect.[24-26] The grain boundary is regarded as a slab like domain, which presents a repulsive potential to the electrons as a shield. Different from the quantum mechanical method KKR used in reference 24-26, we assume there exists spherical metal clusters in electromigration and assume them as hard sphere. We firstly use

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quantum Bernoulli equation combined with spherical flow model to analyze the current-indcued stress caused by electron density gradient, then we analyze the current-indcued stress caused by quantum viscosity. The calculated results agree well with measured values. Then, we calculated the cluster radius and the number of electron per cluster and the calculated results corresponds well with magic numbers, which indicates that there might be large group of atoms transportation during electromigration but not just single-particle-transport.

Quantum density stress (QDS).—Electron fluid is the relative reference frame, and it moves with velocity V ($V = v_e - v_{ato}$) , v_{ato} is the spherical cluster velocity which is 0 in the current direction, v_e is the free electron drift velocity. On the sphere surface, the distribution of relative velocity is $v_{r=a} = \frac{3}{2} V \sin\theta$, where θ is the angle between the radius and x-coordinate.[21] Average the horizontal component of velocity of the left side of sphere, we arrive at

$$\overline{v} = \frac{1}{\pi} \int_{-\frac{\pi}{2}}^{\frac{\pi}{2}} \frac{3}{2} V \sin^2\theta d\theta = \frac{3}{4} V = \frac{3}{4} v_e \qquad (1)$$

Similarly, the velocity of the other side can be got as $\frac{3}{4} v_e$. Assuming that A is at the left side of dislocation and B is at infinity. The mean horizontal velocity of the electron gas at A is v_A, the horizontal velocity of the electron gas at B is v_e, which is the electron gas drift velocity. The pressure of the electron gas at A is P_A. The pressure of the electron gas at B is $P_B = P$ and $\rho_B = \rho$, where P is the mean electron fluid pressure, ρ is the mean electron fluid mass density. Each side of the dislocation atom is assumed to meet Quantum Bernoulli equation with different

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density, respectively. We use the Bernoulli equation for electron fluid developed by M. Di. Ventra[20]

$$\frac{v^2}{2}+\frac{P}{\rho_e}+\frac{V_{ext}}{m_e}=constant \tag{2}$$

where $\rho_e=\rho/m_e$, ρ_e is the electron distribution density, m_e is the electron mass. $P=\rho_e E_f$. For conduction electrons, only those very close to the Fermi energy can participate in the conduction process. Therefore, their pressure is $P=\rho_e E_f$, where E_f is Fermi level. The pressure equation for electron gas at A is obtained as:

$$P_A=\frac{\rho_A}{\rho}P+\rho_A\frac{v_e{}^2-v_A{}^2}{2} \tag{3}$$

Similarly, for the other side C

$$P_C=\frac{\rho_C}{\rho}P+\rho_C\frac{v_e{}^2-v_C{}^2}{2} \tag{4}$$

where $v_C=v_A=\frac{3}{4}v_e$. According to D'Alembert paradox, the pressure and velocity of both sides should be the same. However, the electron gradient between two sides of the cluster in the dislocations line leads to the quantum density stress. Then, we obtain

$$\sigma_{QDS}=P_A-P_C=\frac{\Delta\rho}{\rho}P+\Delta\rho\frac{v_e{}^2-\left(\frac{3}{4}v_e\right)^2}{2}=\frac{\Delta\rho_e}{\rho_e}P+\frac{7}{32}\Delta\rho_e m_e v_e{}^2 \tag{5}$$

For the second part of equation (5), it's in fact the kinetic energy transfer from electrons to ions. The electron fluid flows with velocity v_e and transfers E_f to ions. Therefore, $\frac{1}{2}m_e v_e^2$ should be replaced by $\frac{1}{2}E_f$, where $\frac{1}{2}$ is the factor on average. Then equation (5) can be rewritten as

$$\sigma_{EDF}=\Delta\rho_e E_f+\frac{7}{32}\Delta\rho_e E_f\cong 1.22\Delta\rho_e E_f \tag{6}$$

$\Delta\rho_e$ is the electron distribution density gradient between two sides of clusters.

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The distribution of electron is significantly affected by various factors such as charged vacancies and congregated solute atoms.[27-31] Between the two sides of dislocation, under the effects of charged vacancies, a vivid contrast of electron distribution density is formed. The electron density gradient has been observed in previous researches. The constrictions in current flow path can increase local density up to about a factor of 2.[32] Also, atomic relaxation at step edges redistributed local charge density,[33,34] with enhancement up to a factor of 10 on Cu(532) near kink sites.[35] In reference 36, using DFT calculation, the charge density gradient of Ag can be represented as:

$$\int \delta\rho_e dx = \int (\mu_L - \mu_R)[\rho_L(\mu_{eq}, r) - \rho_R(\mu_{eq}, r)]dx / 2 \qquad (11)$$

And the simulation results indicates that could be of 4×10^{-27}m^{-3} on chain and dislocation lines, which is directly suitable for present model. The conduction electron density ρ_e for single-crystal Ag is 5.85×10^{28} m^{-3}, Fermi energy is 5.48eV, therefore, according to equation (6), $\sigma_{EDF} = 4.25Gpa$ compared with 2GPa which is the current induced stress of Pt thin films.[7] Equation (6) contains two parts. The left part derives from the Fermi pressure, while the other part derives from the quantum kinetic energy transfer. We use classical fluid mechanics to deduce the basic equations for EDF. Then, we further transform it into quantum mechanical forms, and the calculated results demonstrate the success for the transformation we have made.

Quantum viscous Stress (QVS).—In the first part, we consider the electron fluid as ideal fluid and analyze the effect of electron density difference/gradient. In this part, we assume the electron fluid as viscous fluid and analyze the viscosity-induced pressure. The d.c. viscosity of homogeneous electron fluid could be obtained by AK's

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approach[20]

$$\mu = \frac{\rho_e \hbar}{\dot{r}_s^6}\left(\frac{1.81\times10^3}{T}\right)^2\left(\frac{\pi(1+2\dot{r}_s)}{8\sqrt{\dot{r}_s+\dot{r}_s^2}}-\frac{\pi}{4}\right)^{-1} \tag{7}$$

where $\dot{r}_s = \alpha r_s / \pi$ and $\alpha = (4/9\pi)^{1/3} = 0.52$. For analyzing the effect of quantum viscosity, we mainly use hydrodynamics. In classical spherical flow model, under stokes linearization approximation, the 2-dimensional pressure distribution on the sphere surface could be represented as[20]

$$P = P_0 - \frac{3V\mu x}{2R^2} = \rho_e E_f - \frac{3v_e\mu x}{2R^2} \tag{8}$$

where R is the radius of the sphere. The mean pressure on the left side is $P_l = \rho_e E_f + \frac{3v_e\mu}{4R}$. Similarly, the mean pressure of the right side is $P_r = \rho_e E_f - \frac{3v_e\mu}{4R}$. The quantum viscous stress is thus the difference between the two sides.

$$\sigma_{QVS} = \frac{3v_e\mu}{2R} = \frac{3j\hbar}{2eR\dot{r}_s^6}\left(\frac{1.81\times10^3}{T}\right)^2\left(\frac{\pi(1+2\dot{r}_s)}{8\sqrt{\dot{r}_s+\dot{r}_s^2}}-\frac{\pi}{4}\right)^{-1} \tag{9}$$

The quantum viscous stress, in fact, could be influenced by some other carrier scattering mechanisms or lattice vibration. Therefore, we should add a constant K into equation (9) to represent the effective quantum viscous stress. When K=1/2, we find a very interesting result.

TABLE 1. j is the current density, T is the temperature, σ is the previous results of current-induced stress. N is the number of electrons inside each characteristic sphere and $N = \frac{4\pi}{3}\left(\frac{R}{l}\right)^3 Z$ where Z is the number of free electrons per atom. l is the average distance between atoms, R is the radius of metal cluster.

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System	j (10^{10}A/m^2)	T K	σ (MPa)	$\dot{r}_s$	l 10^{-10}m	R (K=1/2) 10^{-10}m	N
Al conductor line (measured)[10]	0.4	534	210	0.34	2.55	3.64	36.55
Al conductor line (calculated)[9]	1	554	510	0.34	2.55	3.49	32.21
Pt thin film (measured)[7]	2.5	350	2000	0.38	2.47	3.25	19.08
Sn solder joint (calculated)[5]	0.14	360	155	0.37	3.00	2.52	9.93

In table 1, the number of electrons inside each cluster of Al, Pt and Sn corresponds to the magic number 34, 20, 8. More surprisingly, the calculation indicates that the 3 different kinds of material, at different temperature, under different current density, in 4 independent experiments,[5,7,9,10] have the same order of magnitude of the cluster radius. This could be a strong corroboration to our assumption which is based on spherical jellium model. According to spherical jellium model, nearspherical metal clusters with magic number are exceptionally strong because of their closed electronic shells.[18,19,23] This might indicate that the electromigration transport should not only be single-particle transport, a cluster of atoms might be transported altogether. It is worth noting that Stokes approximation is satisfied when the Reynolds number $R_e = v_e L_{sys} / \mu \rho_e m_e$ is much smaller than 1. Therefore, we should calculate the value of Reynolds number of electron fluid. According to the value in Table 1, the Reynolds number is much less than 1, even when the characteristic size L_{sys} of the system is $10^{-7} m$. Besides, equation (9) could only provide a very rough estimation of cluster radius, which mainly determines the value of L_{sys}, a more accurate and well correlated approach is needed to get the accurate value of cluster radius.

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The electron viscous fluid could also be applied to explain the motion of nanoislands. Chenggang Tao et al observed the current-biased displacement of monatomic islands of several nanometers on single-crystal Ag.[17] It was found that the velocity of the islands varies inversely with radius, which is contradictory to the prediction of pure attachment/detachment mechanism. However, this phenomenon can be well explained by our model. Assuming the nanoisland is a disk, then the drag force of the disk could be represented as $F_{disk} = KVR_0$,[21] where K is a constant in this system. the island area density is ρ_{is}, the island radius is R_0, the island drift velocity is v_0.

$$\frac{dv_0}{dt} = K\frac{(v_e - v_0)}{\rho_{is}\pi R_0} \tag{10}$$

Then, we can get

$$v_e - Ae^{-\frac{K}{\rho_{is}\pi R_0}t} = v_0 \tag{11}$$

Because v_0=0 when $t = 0$, then $A = v_e$, therefore

$$v_0 = v_e(1 - e^{-\frac{K}{\rho_{is}\pi R_0}t}) \tag{12}$$

$$\frac{\partial v_0}{\partial R_0} = -\frac{Kv_e}{\rho_{is}\pi R_0^2}e^{-\frac{K}{\rho_{is}\pi R_0}t} \tag{13}$$

According to equation (13), the island drift velocity v_0 varies inversely with radius R_0, which well suits the experimental results. Additionally, according to equation (13), the island drift velocity will never be larger than the electron drift velocity. This phenomenon is thus explained in a completely different way from the diffusion mechanism. An important observation was made by K.C. Chen et al.[17] They

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found that the grain boundary slowed down electromigration rate by one order of magnitude. According to (6), the electromigration direction might be opposite to the electron density gradient in grain boundary, which leads to the drag effect.

In reference 37,38, a solution has been obtained for the motion of void driven by electric potential and gradient stress field. If we consider the contribution of QDS and QVS, this chemical potential of the atoms on the interface should be rewritten as

$$\mu = \mu_0 - \Omega\gamma_s\kappa - Z^*eU - \Omega\sigma' - \Omega\sigma_n + \Omega U_s \qquad (14)$$

where μ_0 is the reference value of the potential, γ_s is the interface energy, κ is the curvature of the interface, Ω is the atomic volume, U is the applied electric potential, U_s is the strain energy density of matrix material, σ_n is the normal stress acting on the interface. σ' represents the quantum density pressure and quantum viscous pressure. The atomic flux along the interface is given by

$$J = -\frac{D_i\delta_i}{\Omega kT}\frac{\partial\mu}{\partial s} = \frac{D_i\delta_i}{KT}\left(\gamma_s\frac{\partial\kappa}{\partial s} - Z^*eE_i + \frac{\partial\sigma'}{\partial s} + \frac{\partial\sigma_n}{\partial s} - \frac{\partial U}{\partial s}\right) \qquad (15)$$

where D_i is the interface diffusivity, δ_i is the thickness of the interface, kT has the usual meaning, s is the arc length of the interface, $E_i = -\nabla_s U$. The mass conservation requires that $dJ/dy = V/\Omega$, then integrate it from y to $y = d\sqrt{(1-m)/(1+m)}$, where m is the shape factor, we get

$$J = \frac{V}{\Omega}\left(d\sqrt{\frac{1-m}{1+m}} - y\right) + C \qquad (16)$$

where the constant C can be determined by boundary condition, m is the shape factor, then

$$C = -\frac{D_i\delta_i}{kT}\left(Z^*eE_i + \nabla_s\sigma' + \frac{\partial\sigma_n}{\partial s} - \frac{\partial U}{\partial s}\right) \qquad (17)$$

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Symmetry requires that the flux at $y = 0$ must be zero, thus

$$V = \frac{\Omega D_i \delta_i}{dkT} \sqrt{\frac{1+m}{1-m}} \left(eZ^* E_i + \nabla_s \sigma' - \frac{\partial \sigma_n}{\partial s} + \frac{\partial U}{\partial s} \right) \tag{18}$$

According to equation (18), quantum density stress contributes to the motion of voids. σ' helps explain that the variation of the void velocity should result from the variation of electron density distribution.[3,6] Also, QVS of σ' in equation (15) helps explain the rate of grain boundary diffusion increasing with the current density.[7]

Conclusion.—The current-induced force had been called "electron wind force" for a long time, however, it was merely a concept. In present model, this force is finally demonstrated as a real electron wind. Our model assumes the electron gas as Newton fluid, which is a very bold assumption and completely different from previous researches. However, the calculated results and well explanation of relative phenomenon strongly corroborates our assumption. We calculate the electron force using quantum viscosity, Fermi gas, Stokes drag pressure and Quantum Bernoulli formula, and the calculated value agrees well with experimental value. Our model is also suitable for the motion and force analysis of materials of various scales, such as atoms, nanoislands, dislocations and voids.

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